



RESPONSE UNDER 37 C.F.R. 1.111
U.S. Application 09/305,019

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REMARKS

Entry of the above amendments and reconsideration and withdrawal of the rejections of record are respectfully requested.

Summary of Status of Amendments and Office Action

Claim 4 to 12 are presently pending in the application, with claim 4 being independent. Claim 10 is amended to correct the inadvertent addition of an "n" to the word "ratio" when the claims were submitted with the prior appeal brief. Dependent claims 11 and 12 are being added to include the Si/Al₂ ratio of the synthesis mixture as supported by page 7, line 16, of the specification.

The Examiner has rejected claims 4 to 9 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608, and has rejected claim 10 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608, and further in view of Innes et al., U.S. Patent No. 4,891,458.

Response to the Rejection of Claims 4 to 9

Claims 4 to 9 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608.

In making this rejection, the Examiner stated with respect to independent claim 4:


Cheng discloses alkylation and transalkylation processes in which a feed containing an aromatic compound (e.g., benzene) and olefins (e.g., ethylene) is contacted with MCM-56 or beta zeolite catalyst in an alkylation zone to produce monoalkylbenzene products such as ethylbenzene. By-products (e.g., polyalkylbenzene), which are produced in the alkylation step, can be recycled to the alkylation reactor to undergo transalkylation or they can be reacted with additional aromatic feed in a separate reactor in the presence of TEA-mordenite catalyst to produce monoalkylbenzene products. Cheng discloses reaction conditions for the alkylation step wherein the reaction takes place under liquid phase conditions at temperatures between 150 and 260°C and pressures up to 3000 psig in a space velocity of from

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0.01 to 20 WHSV, based on ethylene feed. Cheng also indicates that the ratios of benzene to ethylene in the alkylation reactor may be from 1:1 to 30:1 molar. Cheng further discloses that the transalkylation zone is operated in a stoichiometric excess of aromatic compounds. Therefore, the ratio of benzene to polyalkylbenzene is greater than one. (See col. 3, lines 25-27; col. 5, lines 19-25; col. 6, lines 1-3; col. 12, line 10 through col. 14, line 40).

The Examiner also rejected dependent claims 5 to 9 over the same combination of references. For purposes of response here, Applicants will direct attention to independent claim 4.

Applicants respectfully submit that there is no *prima facie* case of obviousness, i.e., there is no suggestion to combine the teachings of Kuchenmeister et al. with those of Cheng et al. Further, if a *prima facie* case of obviousness has been established, which it has not, then Applicants have rebutted that case with evidence of unexpected results. 

Cheng et al, column 13, line 40 to column 14, line 44, teach a two step alkylation process utilizing two stages, wherein an aromatic feed (benzene) and olefin are reacted in the presence of an alkylation catalyst (such as MCM-56) to produce polyalkylated product and then reacting the polyalkylated product and additional aromatic feed (benzene) in the presence of "a suitable transalkylation catalyst" (column 14, line 27). "The transalkylation catalyst may be a catalyst comprising a zeolite such as MCM-49, MCM-22, PSH-3, SSZ-25, zeolite X, zeolite Y, zeolite beta, or mordenite," (emphasis added) including a TEA-mordenite, Cheng et al. column 14, lines 27-39. Cheng et al., however, do not set forth the particle size of their transalkylation catalyst, nor do they indicate any preference for the use of a TEA-mordenite. Rather, at best Cheng et al. indicate that any transalkylation catalyst can be utilized. Referring to the claims for guidance with respect to preferences, as would be common for one of ordinary skill in the art, it is noted that dependant claim 3 lists MCM-22, zeolite X, zeolite Y, zeolite beta, and mordenite; dependant claims 6 and 9 require zeolite beta.

It is respectfully submitted that the Kuchenmeister et al. reference must be considered as a whole, and that the teaching of Kuchenmeister et al. is misconstrued by the Examiner's use of a single sentence separate from the context in which that sentence

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occurs. One of ordinary skill in the art would not have been taught what the Examiner is able to see with the advantage of hindsight.

Kuchenmeister et al, page 2, lines 12 to 28, described the problem being solved as excessive production of polyalkylbenzenes at high temperatures and pressures associated with vapor phase reactions. At page 3, lines 19 and 20, Kuchenmeister et al. teach that "the catalyst" used in their invention is selective to the production of ethylbenzene at conditions of about 250 to about 500°C at pressures from about 200 to about 500 psi, giving no indication that such a catalyst would be expected to perform similarly under liquid phase conditions. The statement by Cheng et al. indicating that the transalkylation reaction could be carried out under liquid or vapor phase conditions is not, therefore, applicable to the teachings of Kuchenmeister et al. with respect to a specific catalyst which has been stated to be effective at vapor phase conditions. It is respectfully submitted that the disclosure of Kuchenmeister et al. would actually teach away from using their catalyst or any one of its qualities in a liquid phase reaction. One of ordinary skill in the art would have read the Kuchenmeister et al. disclosure with the understanding that many catalysts effective for promoting vapor phase reactions have not been found to be similarly effective for promoting liquid phase reactions. It is respectfully noted that different catalysts are generally used for vapor and liquid phase reactions because the reaction kinetics and catalyst activity are both affected by the pressure and temperature at which the reaction is occurring. Catalyst properties which improve the results of vapor phase reactions cannot, therefore, be presumed to have a similar effect with respect to liquid phase reactions.

It is respectfully noted that the complete sentence at page 3, lines 29 and 30, in which Kuchenmeister et al. discuss the effect of crystal size is "The improvement in transalkylation control of the process of the present invention is attributed to the use of an aluminosilicate catalyst having a crystal size less than about 0.50µm (0.50 microns)" (emphasis added). The "present invention" has already at that point in the text been defined to include use of a catalyst which is effective at vapor phase conditions, is primarily monoclinic, and preferably has a Si/Al atomic ratio in the range of from about 50 to about 500 (page 3, lines 19 to 22 and 26 to 27). It is respectfully submitted that the context of the crystal size restriction would not have indicated to someone of ordinary skill

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in the art that the crystal size alone of the characteristics taught should be applied to a certain unrelated transalkylation catalyst.

It has long been established that catalysts are generally considered unpredictable merely from the chemical nature of the catalyst. (*Corona Co. v. Dovan* (USSC 1928) 276 US 358, 369). Catalytic effects are not ordinarily predictable with certainty. *In re Doumani et al.* (CCPA 1960) 281 F.2d 215, 126 USPQ 408. Further, the effect of the modification of one prior art catalytic process in a manner employed in another prior art process which employs a different catalyst was held unpredictable. (*Ex parte Berger et al.*, (POBA 1952) 108 USPQ 236). To find obviousness, "there must be some reason for the combination other than the hind sight gleaned from the invention itself." *Interconnect Planning Corp. v. Feil*, 227 U.S.P.Q. 543, 551 (Fed. Cir. 1985). Stated in another way, "[I]t is impermissible to use the claimed invention as an instruction manual or 'template' to piece together the teachings of the prior art so that the claimed invention is rendered obvious." *In re Fritch* 23 U.S.P.Q.2d 1780, 1784 (Fed. Cir. 1992).

It is respectfully noted that there are a multitude of aluminosilicate catalysts and that Kuchenmeister et al. did not provide any enabling direction as to how to synthesize the catalyst they described or even what aluminosilicate catalyst was actually tested. One skilled in the art would not have been likely to put much credence in the indefinite disclosures made. There is no presumptive correlation that two similar processes form substantially the same product where the processes differ by a materially limiting step. *Cf. In re Hoeksema*, 399 F.2d 269, 274, 158 USPO 596, 601 (CCPA 1968) (if the prior art of record failed to disclose a method for making a claimed compound, at the time the invention was made, it cannot be legally concluded that the compound itself was in the possession of the public)

At page 3, lines 19 to 22, Kuchenmeister et al. state that the catalyst is primarily monoclinic aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure. Applicants submit that there is no suggestion to modify Cheng et al. with the crystal size but not the crystal structure of Kuchenmeister et al. Cheng et al. broadly teach that any transalkylation catalyst including TEA-mordenite may be used as a transalkylation catalyst in the Cheng et al. process. Kuchenmeister et al. teach that their transalkylation

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catalyst is primarily monoclinic aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure. Applicants submit that Kuchenmeister et al. teach away from the use of a mordenite catalyst, since mordenite has a wholly orthorhombic crystalline structure and Kuchenmeister et al. place an upper limit of 40% on the percentage of orthorhombic structure which can be contained in the catalyst of their invention. In this regard, the Examiner's attention is directed to Meir and Olson, "Atlas of Zeolite Structure Types," Appendix A, page 144 (Butterworth-Heinemann 1992). This teaching away makes it non-obvious to apply certain characteristics of the Kuchenmeister et al. disclosure and discard others.

Kuchenmeister et al. further place a limit on the silica to alumina ratio, specifying a Si/Al atomic ratio of 50 to 500. The Examiner's attention is directed to Table 1 of the second Declaration by Dr. Wieslaw J. Roth under 37 CFR 1.132 which indicates that the TEA-mordenite of the present invention can have a Si/Al₂ ratio of 34 (inadvertently misstated as Si/Al in Applicants' previous response dated February 14, 2003) which corresponds to a Si/Al atomic ratio of 17. In fact, the disclosed useful silica/alumina (Si/Al₂) ratio for the synthesis mixture of the present invention is in the range from less than 30 to about 90, preferably between about 35 and about 50, corresponding to Si/Al atomic ratios of less than 15 to about 45 and about 17 to about 25, respectively. These disclosed Si/Al atomic ratios outside the range of the teachings of Kuchenmeister et al. further reduce the argument for obviousness of applying any one characteristic of the Kuchenmeister et al. disclosure to the Cheng et al. disclosure.

Kuchenmeister et al, on page 3, lines 43 to 44, further teach that "the sodium content of the catalyst should be maintained at a level less than about 50 ppm in order to facilitate the desired levels of transalkylation" (emphasis added) While sodium content of the catalyst is not specified in the present specification, it is noted that the synthesis mixture disclosed in Example 1 contains a Na₂O/Al₂ molar ratio of 7.3 implying that sodium levels in the synthesized TEA-mordenite would not be insignificant, and certainly not below about 50 ppm. It would be illogical to apply one constraint linked to transalkylation effectiveness but not others. In fact since mordenite and TEA-mordenite are both generally synthesized in a sodium containing solution, one of ordinary skill in the

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art would consider the sodium restriction to teach away from application of the Kuchenmeister et al. ideas to any form of mordenite.

Applicants submit that there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with those of Kuchenmeister et al. As noted in the previously submitted second Declaration by Dr. Wieslaw J. Roth under 37 CFR 1.132, in Example 1 of Kuchenmeister et al., which employs an unspecified aluminosilicate catalyst having an average crystal size of $0.41\mu\text{m}$, the diethylbenzene conversion rate after 1 day on stream is only 11% and that this rate decreases continuously with time on stream until, after 14.6 days, the diethylbenzene conversion rate is 2.8% (an aging rate of about 6%/day). In Comparative Example 1 of Kuchenmeister et al., which again employs an unspecified aluminosilicate catalyst but having an average crystal size of $0.70\mu\text{m}$, the diethylbenzene conversion rate after 1 day on stream is even lower, at 4.6%, but this increases or remains generally constant until 6.7 days on stream. The data in Kuchenmeister et al. suggests that the unspecified aluminosilicate catalyst used therein has extremely low activity for diethylbenzene conversion irrespective of crystal size and that, in the case of at least the small crystal material, the catalyst ages rapidly. For both of these reasons, it would be counter-intuitive for one of ordinary skill in the art to consider the disclosure in Kuchenmeister et al. in seeking to improve the process of Cheng et al., particularly for a commercial application where long run times and hence low aging rates are critical.

If a *prima facie* case of obviousness has been established, which it has not, Applicants have rebutted that case with unexpected results achieved by the use of small crystal TEA-mordenite in the liquid phase transalkylation reaction. In this regard, the Examiner's attention is directed to the second Declaration by Dr. Wieslaw J. Roth under 37 CFR 1.132, which shows the results of testing comparing a catalyst made in accordance with the present invention and having a particle size of less than 0.5 micron with two commercially available mordenite catalysts also having a particle size of less than 0.5 micron.

Dr. Roth's first Declaration presented data showing that TEA-mordenite having an average crystal size of less than 0.5 micron is more active for the transalkylation of

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diisopropylbenzene than TEA-mordenite having an average crystal size of greater than 5 micron. These data were batch data generated in an autoclave and did not provide information as to the aging characteristics of the catalysts.

Table 1 of the second Declaration displays the results of a series of tests comparing the aging of the small crystal TEA-mordenite of the present invention with the aging of two commercially available, conventional mordenite catalysts. The tests were run for 7-14 days at a temperature of 260°C, a pressure of 500 psig, a benzene to polyethylated benzene weight ratio of 3:1 and a WHSV of 5. It will be seen from Table 1 that the small crystal TEA-mordenite of the present invention had an initial diethylbenzene (DEB) conversion activity of 77%, significantly and surprisingly higher than that of the Kuchenmeister et al. catalysts, but essentially the same as that of the conventional mordenite catalyst supplied by PQ Corporation. However, although each of the catalysts had a crystal size of below 0.5 μm , the small crystal TEA-mordenite of the present invention exhibited an aging rate significantly lower than either of the conventional mordenite catalysts. This surprising reduction of catalyst aging has significant commercial benefit.

The Examiner noted that the three catalysts shown in Table 1 of the second declaration had different silica alumina ratios, alpha values, and surface areas and stated that it was unclear if the aging rate was dependant on one or a combination of these variables. It is respectfully noted that the Applicants are not claiming a reduction in aging based on a specific variable, but have satisfied the requirement of showing that the claimed TEA-mordenite with a crystal size of less than 0.5 micron is surprisingly better than the 0.5 micron crystal size catalyst disclosed by Kuchenmeister et al. Applicants have further shown that even with respect to mordenite type catalysts, the 0.5 micron crystal size TEA-mordenite claimed in the present invention is commercially advantageous over other forms of mordenite available in a small crystal size.

For the foregoing reasons, Applicants respectfully request that the rejection be withdrawn.

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Response to the Rejection of Claim 10

Claim 10 is rejected under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024, in view of Kuchenmeister et al., EP 733,608, and further in view of Innes et al., U.S. Patent No. 4,891,358.

It is Applicants' position that this rejection fails for the same reasons as given above with respect to the rejection of claims 4 to 9, that there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with those of Kuchenmeister et al. If a *prima facie* case of obviousness has been made for the purposes of this rejection, Applicants rely upon the arguments made above with respect to the rejections of claims 4 to 9 and incorporate those reasons herein. For the foregoing reasons, Applicants respectfully request that this rejection be withdrawn.

CONCLUSION

Applicants respectfully submit that the foregoing arguments place the application in condition for allowance. Allowance of this application is therefore earnestly solicited.

Respectfully submitted,

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